

The single-ion anisotropy in LaFeAsO

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Abstract. We use Green's function method to study the Heisenberg model of LaFeAsO with the striped antiferromagnetic collinear spin structure. In addition to the intra-layer spin couplings J_{1a}, J_{1b}, J_2 and the inter-layer coupling J_c , we further consider the contributions of the single-ion anisotropy J_s . The analytical expressions for the magnetic phase-transition temperature T_N and the spin spectrum gap Δ are obtained. According to the experimental temperature $T_N = 138\text{K}$ and the previous estimations of the coupling interactions, we make a further discussion about the magnitude and the effects of the single-ion anisotropy J_s . We find that the magnitudes of J_s and J_c can compete. The dependences of the transition temperature T_N , the zero-temperature average spin and the spin spectrum gap on the single-ion anisotropy are investigated. We find they both increase as J_s increases. The spin spectrum gap at low temperature $T \rightarrow 0$ is calculated as a function of J_s , the result of which is a useful reference for the future experimental researches.

1. Introduction

It was recently discovered that an iron-based material LaFeAsO shows high-temperature superconductivity when O atoms are partially substituted by F atoms[1]. This discovery has triggered great research interest on the FeAs-based pnictides superconductors and their undoped compounds. It has been theoretically and experimentally confirmed that these pure FeAs-based compounds have a ground state with collinear stripe-like antiferromagnetic(AF) spin order formed by Fe atoms[2, 3, 4, 5, 6, 7, 8, 9, 10, 11]. Thus to establish an effective spin Hamiltonian for them and to elucidate the corresponding antiferromagnetism are helpful in understanding the underlying mechanism to make them superconducting upon doping.

For undoped LaFeAsO and other similar parent compounds, a Heisenberg exchange model was suggested to explain their AF structure[2, 4, 12, 13], and was used to explore their magnetic properties[14, 15, 16]. Figure 1 shows the unit cell of the orthorhombic AF spin structure of the Fe lattice. This orthorhombic structure exists below a structure transition temperature T_S , which is 15 ~ 20K higher than the magnetic transition temperature T_N [6, 17]. Usually the nearest neighbor (NN) coupling J_1 (including J_{1a} and J_{1b}), and the next-nearest neighbor (NNN) coupling J_2 in FeAs layers are dominant and must be considered. The NN coupling J_c between spins on neighboring layers is regarded to be much smaller than the in-plane couplings[2, 4]. However, J_c was found to be essential for the existence of a non-zero magnetic transition temperature T_N [15]. A further consideration can include the single-ion anisotropy J_s . It was estimated to be even much smaller than J_c in the model of SrFe₂As₂, but a spin spectrum gap was found to be produced by it[14]. For LaFeAsO, so far there is no research report about the magnitude or the effects of the single-ion anisotropy.

In this paper, we use Green's function method[18] to study the Heisenberg model of FeAs-based pure parent compounds. The Hamiltonian of this model in a detailed form is

$$H = \frac{1}{2}J_{1a} \sum_{\langle ij \rangle} \mathbf{S}_{1i} \cdot \mathbf{S}_{1j} + \frac{1}{2}J_{1a} \sum_{\langle ij \rangle} \mathbf{S}_{2i} \cdot \mathbf{S}_{2j} + J_{1b} \sum_{\langle ij \rangle} \mathbf{S}_{1i} \cdot \mathbf{S}_{2j} \\ + J_2 \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_{1i} \cdot \mathbf{S}_{2j} + J_c \sum_{\langle ij \rangle} \mathbf{S}_{1i} \cdot \mathbf{S}_{2j} - J_s \sum_i [(S_{1i}^z)^2 + (S_{2i}^z)^2], \quad (1)$$

where the spin coupling J_c between layers and the single-ion anisotropy J_s are both considered. The subscripts 1 and 2 mean the sublattices 1 and 2 respectively. $\langle ij \rangle$ means NN spin pairs, and $\langle\langle ij \rangle\rangle$ means NNN spin pairs. The self-consistent equations for the average sublattice spin will be derived. An analytical expression for the magnetic transition temperature T_N will be obtained. For LaFeAsO, according to the recent estimations of the strengths J_{1a} , J_{1b} , J_2 and J_c [15] with the experimental temperature $T_N = 138\text{K}$ [6, 17], we shall make a further estimation of the single-ion anisotropy J_s . We find that the magnitude of J_s can compete with J_c , and in some situations even bigger than J_c . The effects of the single-ion anisotropy on the transition temperature T_N , the zero-temperature average spin $\langle S_z \rangle_0$ and the spin spectrum gap are investigated. We

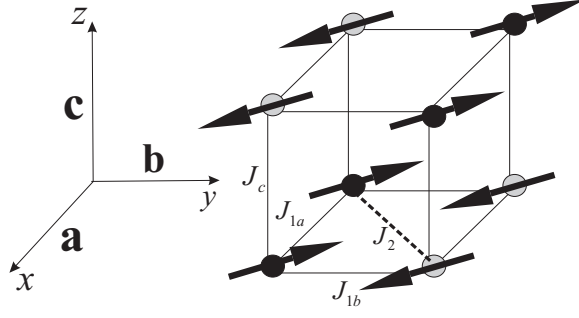


Figure 1. A unit cell of the orthorhombic Fe spin lattice. \mathbf{a} , \mathbf{b} , \mathbf{c} are the three base vectors. The lattice consists of two sublattices (distinguished by the color gray and black).

find they all increase as J_s increases. In section 2, we shall give our analytical results derived from the Green's function method. In section 3, we shall present our numerical results. Finally a conclusion is given in section 4.

2. Green's function derivation

According to the general scheme of Green's function method to solve an antiferromagnetic spin model with two sublattices, we construct the following Green's functions:

$$G_{1k,1l}(\omega) = \langle\langle S_{1k}^+; S_{1l}^- \rangle\rangle, \quad G_{2k,1l}(\omega) = \langle\langle S_{2k}^+; S_{1l}^- \rangle\rangle. \quad (2)$$

The equation of motion is

$$\omega \langle\langle A; S_{1l}^- \rangle\rangle = \langle[A, S_{1l}^-] \rangle + \langle\langle [A, H]; S_{1l}^- \rangle\rangle, \quad (3)$$

where A represents the spin operator S_{1k}^+ or S_{2k}^+ . The commutator $[A, H]$ can be derived using Hamiltonian (1) and the basic commutation relations of spin operators: $[S_i^+, S_j^-] = 2S_i^z \delta_{ij}$, $[S_i^z, S_j^\pm] = \mp S_i^\pm \delta_{ij}$, where $S_i^\pm = S_i^x \pm iS_i^y$.

In order to close the system of equations, the so-called PRA or Tyablikov decoupling[18] is adopted for the terms stemming from the exchange couplings:

$$\langle\langle S_i^z S_j^+; S_l^- \rangle\rangle \approx \langle S_i^z \rangle \langle\langle S_j^+; S_l^- \rangle\rangle, \quad i \neq j. \quad (4)$$

While for the terms stemming from the single-ion anisotropy, we adopt the Anderson-Callen(AC) decoupling[19]:

$$\langle\langle S_i^z S_i^+ + S_i^+ S_i^z; S_j^- \rangle\rangle \approx 2\langle S_i^z \rangle \Theta_i^{(z)} \langle\langle S_i^+; S_j^- \rangle\rangle, \quad (5)$$

where

$$\Theta_i^{(z)} = 1 - \frac{1}{2S^2} [S(S+1) - \langle S_i^z S_i^z \rangle]. \quad (6)$$

The AC decoupling has been demonstrated to be most adequate for the single-ion anisotropy much small compared to the exchange interactions[20, 21].

In order to write the decoupled equations of motion in the \mathbf{k} space, we take the following Fourier transformation:

$$G_{k,l}(\omega) = \frac{1}{N} \sum_{\mathbf{k}} G(\mathbf{k}, \omega) e^{i\mathbf{k} \cdot (\mathbf{R}_k - \mathbf{R}_l)}, \quad (7)$$

where N is the number of sites in either sublattice, and the summation over \mathbf{k} is restricted to the first Brillouin zone of the sublattice. At the same time, the equation $\delta_{ij} = \frac{1}{N} \sum_{\mathbf{k}} e^{i\mathbf{k} \cdot (\mathbf{R}_i - \mathbf{R}_j)}$ is also used.

Because of the translation invariant, we have $\langle S_{1k}^z \rangle = \langle S^z \rangle$, $\langle S_{2k}^z \rangle = -\langle S^z \rangle$ and $\Theta_{1k}^{(z)} = \Theta_{2k}^{(z)} = \Theta^{(z)} = 1 - \frac{1}{2S^2} [S(S+1) - \langle S^z S^z \rangle]$. Finally, we obtain the decoupled equations of the two Green's function in \mathbf{k} space:

$$[\omega - \langle S^z \rangle A_{\mathbf{k}}] G_{11}(\mathbf{k}, \omega) - \langle S^z \rangle B_{\mathbf{k}} G_{21}(\mathbf{k}, \omega) = 2\langle S^z \rangle, \quad (8)$$

and

$$[\omega + \langle S^z \rangle A_{\mathbf{k}}] G_{21}(\mathbf{k}, \omega) + \langle S^z \rangle B_{\mathbf{k}} G_{11}(\mathbf{k}, \omega) = 0, \quad (9)$$

where

$$A_{\mathbf{k}} = 2J_{1a} \cos(k_x a) - 2J_{1a} + 2J_{1b} + 4J_2 + 2J_c + 2J_s \Theta^{(z)}, \quad (10)$$

and

$$B_{\mathbf{k}} = 2J_{1b} \cos(k_y b) + 4J_2 \cos(k_x a) \cos(k_y b) + 2J_c \cos(k_z c), \quad (11)$$

in which a, b, c are the three lattice constants. Solving equations (8) and (9), we obtain the Green's function:

$$G_{11}(\mathbf{k}, \omega) = \frac{\langle S^z \rangle}{\omega_{\mathbf{k}}} \left[\frac{A_{\mathbf{k}} \langle S^z \rangle + \omega_{\mathbf{k}}}{\omega - \omega_{\mathbf{k}}} - \frac{A_{\mathbf{k}} \langle S^z \rangle - \omega_{\mathbf{k}}}{\omega + \omega_{\mathbf{k}}} \right], \quad (12)$$

and the spin spectrum:

$$\omega_{\mathbf{k}} = \langle S^z \rangle \sqrt{A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2}. \quad (13)$$

When $\mathbf{k} \rightarrow 0$, we obtain a expression for the spectrum gap:

$$\Delta = 2\langle S^z \rangle \sqrt{J_s \Theta^{(z)} [2J_{1b} + 4J_2 + 2J_c + J_s \Theta^{(z)}]}, \quad (14)$$

which is similar with the expression given in ref[14] derived from the spin-wave theory, expect for the factor $\Theta^{(z)}$. From this expression for the gap, one see that the single-ion anisotropy is essential for the existence of the spectrum gap.

Then following the process of solving the average spin, we derive the correlation function $\langle S^- S^+ \rangle$ using the spectrum theorem:

$$\begin{aligned} \langle S^- S^+ \rangle &= -\frac{1}{N\pi} \sum_{\mathbf{k}} \int_{-\infty}^{\infty} d\omega \frac{\text{Im} G_{11}(\mathbf{k}, \omega + i\epsilon)}{e^{\beta\omega} - 1} \\ &= \frac{\langle S^z \rangle}{N} \sum_{\mathbf{k}} \left[\frac{A_{\mathbf{k}}}{\sqrt{A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2}} \coth \frac{\beta\omega_{\mathbf{k}}}{2} - 1 \right], \end{aligned} \quad (15)$$

in which the equation $\frac{1}{x+i\epsilon} = P(\frac{1}{x}) - i\pi\delta(x)$ ($P(\dots)$ means taking the principle value) has been used to obtain the imaginary part of $G_{11}(\omega + i\epsilon)$, and $\beta = \frac{1}{k_B T}$, k_B is the Boltzmann constant, T is the temperature.

According to the theory of Callen[22], the average spin for arbitrary S can be calculated using the following equation:

$$\langle S^z \rangle = \frac{(S - \Phi)(1 + \Phi)^{2S+1} + (S + 1 + \Phi)\Phi^{2S+1}}{(1 + \Phi)^{2S+1} - \Phi^{2S+1}}, \quad (16)$$

where

$$\begin{aligned} \Phi &= \frac{\langle S^- S^+ \rangle}{2\langle S^z \rangle} \\ &= \frac{1}{2N} \sum_{\mathbf{k}} \left[\frac{A_{\mathbf{k}}}{\sqrt{A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2}} \coth \frac{\beta \omega_{\mathbf{k}}}{2} - 1 \right]. \end{aligned} \quad (17)$$

On the other hand, the correlation function $\langle S^z S^z \rangle$ can be calculated from the equation $\langle S^z S^z \rangle = S(S + 1) - (1 + 2\Phi)\langle S^z \rangle$. Using equation (6), we can relate $\Theta^{(z)}$ to Φ by

$$\Theta^{(z)} = 1 - \frac{\langle S^z \rangle}{2S^2} (1 + 2\Phi). \quad (18)$$

Now the equations (16)(17)(18) can be solved self-consistently to obtain the average spin at any given temperature, provided we know the values of the exchange couplings J_{1a}, J_{1b}, J_2, J_c and the single-ion anisotropy J_s .

When the temperature T approaches zero, we obtain $\coth(\frac{\beta \omega_{\mathbf{k}}}{2}) \rightarrow 1$. The equation (17) is reduced to

$$\Phi|_{T \rightarrow 0} = \frac{1}{2N} \sum_{\mathbf{k}} \left[\frac{A_{\mathbf{k}}}{\sqrt{A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2}} - 1 \right]. \quad (19)$$

The zero-temperature average spin $\langle S^z \rangle_0$ can be obtained by self-consistently solving the equations (16)(18)(19).

When the temperature T approaches the magnetic transition temperature T_N , the average spin $\langle S^z \rangle$ as well as the spectrum $\omega_{\mathbf{k}}$ will approach zero. Expanding $\coth(\frac{\beta \omega_{\mathbf{k}}}{2})$ in the equation (17), we obtain

$$\Phi|_{T \rightarrow T_N} \approx \frac{\Gamma}{\beta \langle S^z \rangle} - \frac{1}{2}, \quad (20)$$

where $\Gamma = \frac{1}{N} \sum_{\mathbf{k}} \frac{A_{\mathbf{k}}}{A_{\mathbf{k}}^2 - B_{\mathbf{k}}^2}$. Inserting (20) into (16), and expanding the terms in the denominator and the numerator as the series of $\langle S^z \rangle$, we finally derive

$$\langle S^z \rangle \approx \sqrt{\frac{12(\Gamma k_B T_N)^2}{S(2S - 1)}} \left(1 - \frac{T}{T_N} \right), \quad (21)$$

where

$$T_N = \frac{S(S + 1)}{3k_B \Gamma}. \quad (22)$$

On the other hand, inserting (20) into (18), and using the equation (22), we obtain the reduced expression for $\Theta^{(z)}$ near the temperature T_N :

$$\Theta^{(z)}|_{T \rightarrow T_N} \approx \frac{2S - 1}{3S}. \quad (23)$$

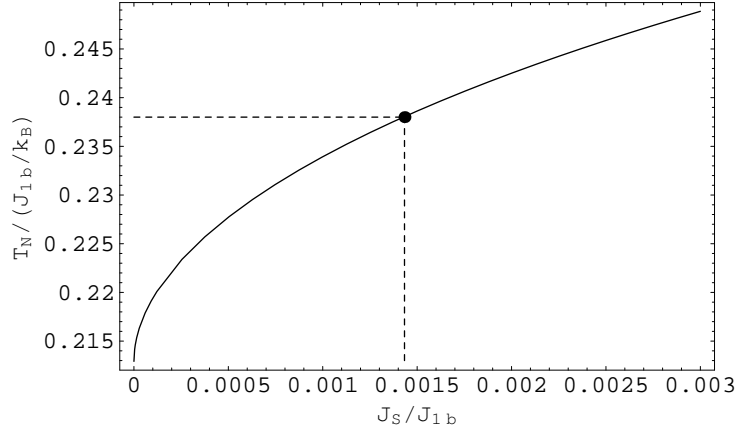


Figure 2. The transition temperature T_N as a function of the single-ion anisotropy J_s for $J_{1a} = 0.98$, $J_2 = 0.52$ and $J_c = 0.0004$ in the unit of $J_{1b} = 50$ meV. The black point correspond to the experimental temperature $T_N = 0.238$ in the unit of J_{1b}/k_B .

3. Numerical results and discussions

So far there is no consensus on the magnitudes of the exchange couplings J_{1a} , J_{1b} , J_2 and J_c , because of the unclear microscopic origin of the observed AF spin structure. Here we prefer the estimations in ref[15], which gave $J_{1b} = 50 \pm 10$ meV, $J_{1a} = 49 \pm 10$ meV, $J_2 = 26 \pm 5$ meV and $J_c = 0.020 \pm 0.015$ meV by using the experimental transition temperature $T_N = 138$ K of pure LaFeAsO. The main purpose of this paper is to investigate the magnitude and the effects of the single-ion anisotropy J_s in LaFeAsO. Through out our numerical calculation, we take $J_{1b} = 50$ meV, $J_{1a} = 49$ meV, $J_2 = 26$ meV and the spin $S = 1$. The result $J_{1b} \sim 50$ meV are obtained from the first-principle calculating[4, 23]. In the present systems of units, the Boltzmann constant is taken as $k_B = 0.086$ meV/K.

Figure 2 shows the effect of the single-ion anisotropy J_s on the transition temperature T_N . We can see that T_N increases as J_s increases. This means that the single-ion anisotropy term is in favor of the AF spin structure. It can be understood from the expression of the single-ion anisotropy term in the Hamiltonian (1). Increasing the magnitude of J_s will make the spins incline to align along the z axis, and give a lower total energy, which make the system more stable. To one's surprise, the magnitude of J_s corresponding to the experimental transition temperature T_N is about $0.00143J_{1b}$, which is much bigger than the magnitude of the exchange coupling $J_c = 0.0004J_{1b}$ estimated in ref[15]. Furthermore, we see from figure 3 that the variation range of $\langle S^z \rangle_0$ with J_s varying from 0 to 0.1 is almost the same as the one produced by J_c in ref[15]. All these results imply that the magnitude of J_s is probably not much small compared with J_c . So the estimation of J_c maybe need to be adjusted if the single-ion anisotropy term is considered. As to the estimations of the other exchange couplings J_{1a} , J_{1b} and J_2 , we think there are still reasonable.

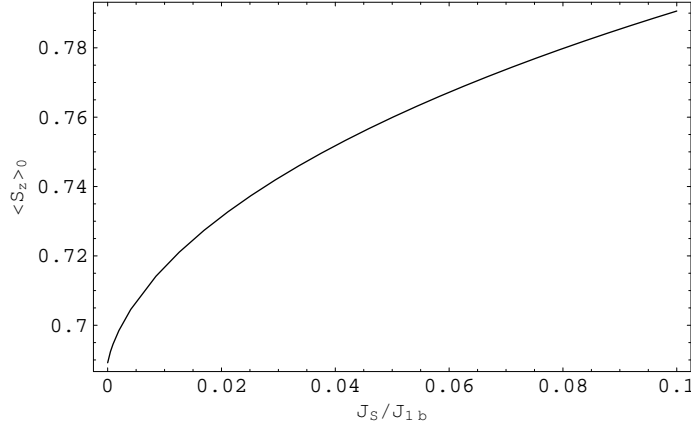


Figure 3. The zero-temperature average spin as a function of the single-ion anisotropy J_s for $J_{1a} = 0.98$, $J_2 = 0.52$ and $J_c = 0.0004$ in the unit of $J_{1b} = 50$ meV.

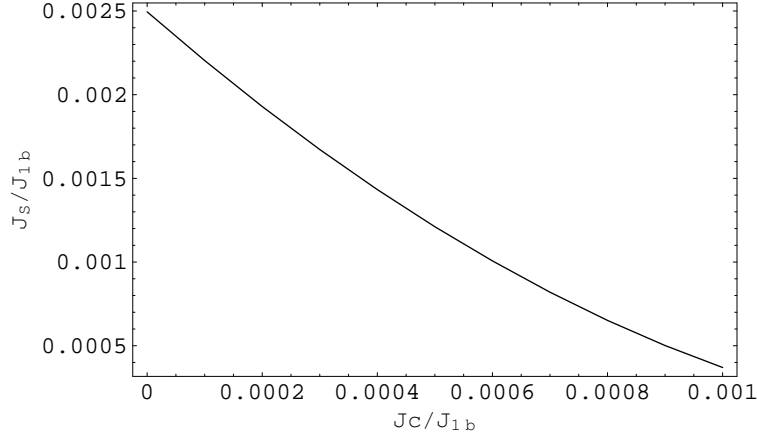


Figure 4. The competing relation of J_c and J_s . The curve is depicted by using the equation (22) for $T_N = 0.238(J_{1b}/k_B)$, $J_{1a} = 0.98J_{1b}$ and $J_2 = 0.52J_{1b}$.

Figure 4 shows the competing relation of J_c and J_s when the transition temperature is fixed at the experimental value. The increase of J_c is accompanied by the decrease of J_s , and vice versa. From figure 4, we can see that the ranges of their corresponding variations are at the same magnitude, which implies they probably have the same status in the viewpoint of theoretical study. As to revealing the actual magnitudes of the two parameters J_c and J_s , we think it is not enough to use only the experimental transition temperature T_N .

Figure 5 shows the effect of the single-ion anisotropy J_s on the spectrum gap at low temperature. The gap vanishes as J_s vanishes, and increases as J_s increases. We find the effect of J_c on the gap is very trivial. The curves for different values of J_c between (0.0001, 0.01) are almost the same, while the single-ion anisotropy affects the gap apparently. Considering the gap can be obtained from inelastic neutron-scattering

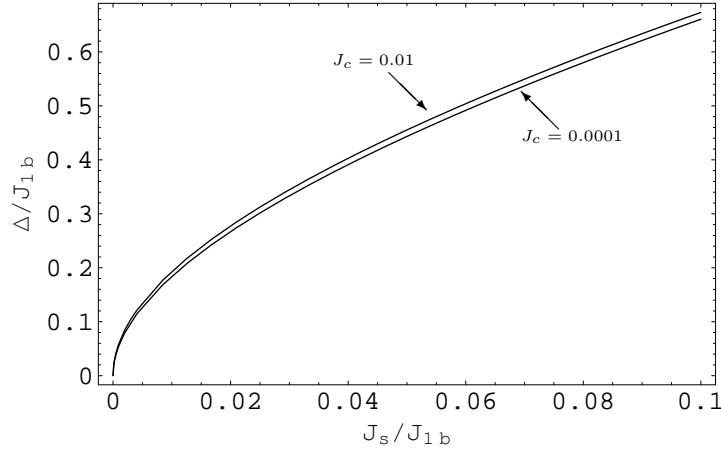


Figure 5. The spin spectrum gaps at low temperature $T \rightarrow 0$ as functions of the single-ion anisotropy J_s for $J_{1a} = 0.98J_{1b}$, $J_2 = 0.52J_{1b}$ and $J_c = 0.0001J_{1b}$ (the below curve), $J_c = 0.01J_{1b}$ (the above curve).

experiment[14], we suggest that the magnitude of the single-ion anisotropy be estimated from the future experimental results of the spectrum gap. For example, if $J_c = 0.0004J_{1b}$, we obtain $J_s = 0.00143J_{1b}$ from the experimental transition temperature $T_N = 138\text{K}$. Then calculating the spectrum gap with $J_s = 0.00143J_{1b}$, we obtain the magnitude of the gap $\Delta \approx 3.4 \text{ meV}$, which can be compared with the future experimental result.

4. Conclusion

We use Green's function method to study the Heisenberg model (1) of LaFeAsO with the striped AF spin structure as shown in figure 1. The main purpose of this paper is to investigate the magnitude and the effects of the single-ion anisotropy J_s . We derive the self-consistent equations for the average spin, and obtained the analytical expressions for the spin spectrum gap Δ , and the magnetic transition temperature T_N . We find that the transition temperature T_N , the zero-temperature average spin $\langle S^z \rangle_0$ and the spin spectrum gap Δ are all increasing functions of the single-ion anisotropy J_s . From our numerical results by using $T_N = 138\text{K}$ and the previous estimations of J_{1a} , J_{1b} , J_2 and J_c in ref[15], we find that the magnitude of J_s is probably not much small compared with J_c . Because the single-ion anisotropy is essential for the existence of the spin spectrum gap, we suggest using the experimental result of the spin spectrum gap to fix the magnitude of the single-ion anisotropy J_s in the future.

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